

ELECTRONIC DENSITY OF STATES FOR INCOMMENSURATE LAYERS

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ABSTRACT. We prove that the electronic density of states (DOS) for 2D incommensurate layered structures, where Bloch theory does not apply, is well-defined as the thermodynamic limit of finite clusters. In addition, we obtain an explicit representation formula for the DOS as an integral over local configurations.

Next, based on this representation formula, we propose a novel algorithm for computing electronic structure properties in incommensurate heterostructures, which overcomes limitations of the common approach to artificially strain a large supercell and then apply Bloch theory.

1. INTRODUCTION

Bloch theory provides an elegant solution for describing the electronic structure of periodic materials. However, there has been a lot of focus recently on the study of *incommensurate* layers of two-dimensional crystal structures [16, 17]. In the absence of periodicity, computing the electronic structure of such materials becomes more challenging.

A common approach to approximate the electronic properties of such a system is to artificially strain it to obtain periodicity on a large supercell, and then apply Bloch theory to this periodic system [4, 9–11, 16]. Commensurate approximations to an incommensurate system are computationally expensive, and their approximation error is unclear. Here we introduce a new method for computing a class of observables derived from the density of states for multi-layer incommensurate heterostructures *without* requiring an artificial strain in the system.

To approximate an observable of an infinite incommensurate system, we approximate local lattice site contributions to the observable. We observe that a site is uniquely defined by its local geometry. Using an equidistribution theorem, there is a predictable distribution of local geometries, and hence site contributions. Consequently, we can express observables in incommensurate heterostructures in terms of an integral over a unit cell, in a fashion rather similar to Bloch theory. This unit cell classification of local configurations is related to Bellisard’s noncommutative Brillouin Zone for aperiodic solids [1]. Prodan used the Bellisard formalism to compute electronic properties for periodic materials with on-site defects modeled by a tight-binding model [13]. Here we consider the density of states and related observables for incommensurate multi-layers.

While the methodology is in principle generic, our derivation and analysis focuses on tight-binding models, which are commonly employed for computing the electronic

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structure of 2D materials [2, 8]. We consider the density of states and related observables for incommensurate multi-layers. We use Chebyshev Polynomial methods to approximate the density of states as a function [6, 12, 14, 15, 19], and from this function any observable can be computed.

Outline. In Section 2 we introduce the results for the bilayer case, and briefly discuss their extension to the multi-layer case. In Section 2.1 we introduce incommensurate systems and the equidistribution result. In Section 2.2 we specify the details of our model problem, and in Section 2.3 we show how to compute the local density of states. In Section 2.4 we prove the infinite system is well posed and express the observables as an integral over local observables.

Section 3 we describe an approximation scheme and present numerical results. In Section 3.1 we discuss the integral discretization. In Section 3.2, we introduce a Chebyshev Kernel Polynomial Method, and in Section 3.4 we present numerical results. In Section 4 we present the details of the proofs.

2. MAIN RESULTS

2.1. Incommensurate Heterostructures. Consider two periodic atomic sheets in parallel 2D planes separated by a constant distance. Each individual sheet can be described as a Bravais lattice embedded in \mathbb{R}^2 by neglecting the out of plane distance. This coordinate is not relevant for classifying the aperiodicity and will be incorporated in section § 2.2. For sheet $j \in \{1, 2\}$, we define the Bravais lattice

$$\mathcal{R}_j = \{A_j n : n \in \mathbb{Z}^2\},$$

where A_j is a 2×2 invertible matrix. We define the *unit cell* for sheet j as

$$\Gamma_j = \{A_j \alpha : \alpha \in [0, 1)^2\}.$$

Each individual sheet is trivially periodic, since

$$\mathcal{R}_j = A_j n + \mathcal{R}_j \quad \text{for } n \in \mathbb{Z}^2.$$

However, the combined system $\mathcal{R}_1 \cup \mathcal{R}_2$ need not be periodic (Figure 1a). (Note that here $\mathcal{R}_1 \cup \mathcal{R}_2$ is only considered to describe geometry, not as an indexing of the atoms as it would have the failure of identifying the origins from each lattice.)

Since we are interested in aperiodic systems, we make the following standing assumption:

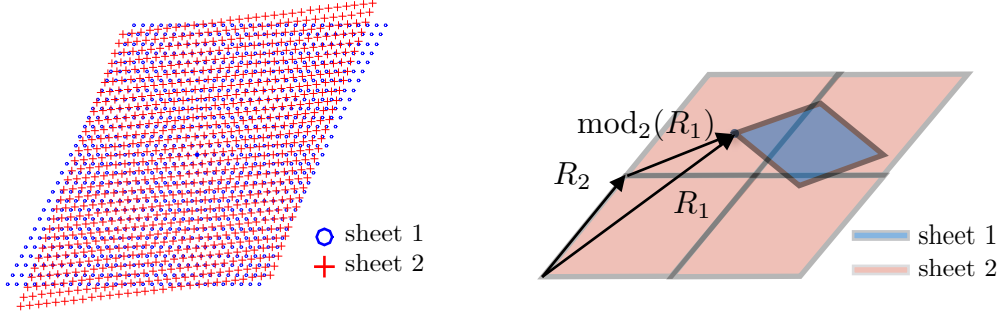
Assumption 2.1. *The lattices \mathcal{R}_1 and \mathcal{R}_2 are incommensurate, that is, for $v \in \mathbb{R}^2$,*

$$v + \mathcal{R}_1 \cup \mathcal{R}_2 = \mathcal{R}_1 \cup \mathcal{R}_2 \Leftrightarrow v = \begin{pmatrix} 0 \\ 0 \end{pmatrix}.$$

Since the majority of material simulation tools rely on periodicity, the most common method at present to simulate incommensurate layers is to adjust one of the two layers slightly in order to make the system commensurate on some larger supercell (Figure 2). In contrast we take advantage of an equidistribution of local geometries.

To parameterize the local geometries, we define the modulation operator $\text{mod}_j : \mathbb{R}^2 \rightarrow \Gamma_j$ on sheet j for position $u \in \mathbb{R}^2$:

$$\text{mod}_j(u) := u + R_j \text{ where } R_j \in \mathcal{R}_j \text{ such that } u + R_j \in \Gamma_j.$$



(A) An incommensurate hexagonal bilayer. Sheet 1 is rotated by $\theta = 6^\circ$ relative to sheet 2. (B) $\text{mod}_2(R_1)$ is the shift of the first lattice relative to the second lattice.

FIGURE 1. Visualisation of incommensurate bilayer geometry.

Then the relative shift of site $R_1 \in \mathcal{R}_1$ is $\text{mod}_2(R_1) \in \Gamma_2$ (See Figure 1b). The local geometry of site $R_1 \in \mathcal{R}_1$ is defined by

$$\mathcal{R}_1 \cup \mathcal{R}_2 - R_1 = \mathcal{R}_1 \cup (\mathcal{R}_2 - R_1) = \mathcal{R}_1 \cup (\mathcal{R}_2 - \text{mod}_2(R_1)).$$

Hence, the local geometry is determined by the relative shift $\text{mod}_2(R_1)$. The same argument holds for relative configurations around a site on sheet two. A fundamental idea in this method is that the distribution of $\text{mod}_j(R_{P_j}) \in \Gamma_j$ is uniform in the sense of Theorem 2.1 below.

We let

$$B_r = \{y \in \mathbb{R}^2 : |y| < r\}, \quad \text{for } r > 0.$$

For $j \in \{1, 2\}$, we let P_j be the transposition, that is, $P_1 = 2$ and $P_2 = 1$.

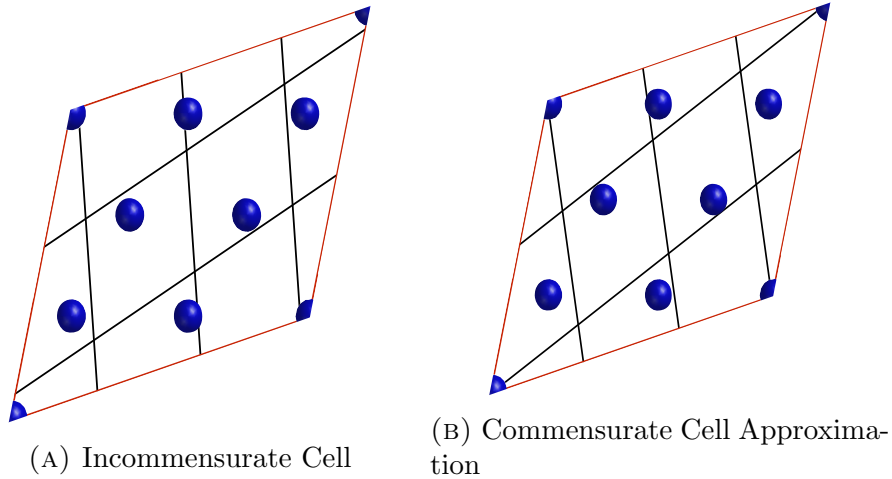


FIGURE 2. (A) Two lattices (spheres and lines) that are incommensurate; (B) The sphere lattice is slightly rotated to obtain a commensurate cell approximation.

Theorem 2.1. *Consider \mathcal{R}_1 and \mathcal{R}_2 incommensurate lattices embedded in \mathbb{R}^2 (i.e., satisfying Assumption 2.1). Then for $g \in C_{\text{per}}(\Gamma_{P_j})$, we have*

$$(2.1) \quad \frac{1}{\#\mathcal{R}_j \cap B_r} \sum_{\ell \in \mathcal{R}_j \cap B_r} g(\ell) \rightarrow \frac{1}{|\Gamma_{P_j}|} \int_{\Gamma_{P_j}} g(b) db.$$

In particular, local geometries around sheet 1 sites can be parameterized by Γ_2 , while local geometries around sheet 2 sites can be parameterized by Γ_1 .

Theorem 2.1 suggests the following strategy for defining *and computing* electronic structure properties in incommensurate heterostructures: (1) Split an observable into local contributions from each atomic site (we will employ the local density of states); (2) Employ Theorem 2.1 to demonstrate that the thermodynamic limit from finite clusters exist (observe that (2.1) is a sum over a finite cluster); (3) Use the right-hand side of (2.1) to compute the limit quantity.

2.2. Tight-Binding Model. Electronic structure is governed by solutions to the Schrödinger eigenproblem. It is typically approximated using methods such as the Kohn–Sham DFT (KS-DFT) model or the Hartree–Fock approximation [8]. For systems in the thousands of atoms however, the standard KS-DFT calculation becomes intractable. The tight-binding (TB) model applies further approximations, and as a result can treat larger systems ranging in the millions of atoms.

Let \mathcal{A}_i denote the set of indices of orbitals associated with each unit cell of sheet i . We assume that \mathcal{A}_i are finite and that $\mathcal{A}_1 \cap \mathcal{A}_2 = \emptyset$. Then the full degree of freedom space is

$$\Omega = (\mathcal{R}_1 \times \mathcal{A}_1) \cup (\mathcal{R}_2 \times \mathcal{A}_2).$$

The interaction between orbitals indexed by $R\alpha$ and $R'\alpha'$ is denoted by $h_{\alpha\alpha'}(R - R')$, where $h_{\alpha\alpha'} \in C(\mathbb{R}^2)$. Although the sheets have a vertical displacement between them, this distance is constant and hence can be encoded into $h_{\alpha\alpha'}$ (using the assumption that $\mathcal{A}_1 \cap \mathcal{A}_2 = \emptyset$). We will further use the following assumption:

Assumption 2.2. *Orbital interactions $h_{\alpha\alpha'}$ are uniformly continuous on \mathbb{R}^2 and decay exponentially, that is,*

$$|h_{\alpha\alpha'}(x)| \leq C e^{-\tilde{\gamma}|x|} \quad \text{for } x \in \mathbb{R}^2.$$

This applies in most scenarios, since in most tight-binding models the orbitals are *tightly bound* around the atomic sites [8], or are exponentially decaying. We then formally define a matrix H such that

$$H_{R\alpha, R'\alpha'} = h_{\alpha\alpha'}(R - R').$$

This is an infinite matrix, hence the eigenproblem

$$H\psi = E\psi$$

for $\psi \in \mathbb{C}^{\mathbb{N}}$ cannot be solved directly. Instead, we will define a class of observables for the infinite system by first defining them for finite sub-systems and then passing to the limit in § 2.4.

For $\tilde{\Omega} \subset \Omega$ with $\#\tilde{\Omega} = n$ the associated hamiltonian is $\tilde{H} = (H_{ij})_{i,j \in \tilde{\Omega}} \in M_n(\mathbb{C})$, where $M_n(\mathbb{C})$ denotes the set of $n \times n$ Hermitian matrices over \mathbb{C} . The *density of states* for $\tilde{\Omega}$ can be defined via its action on test functions, or, observables g , by

$$\mathcal{D}[\tilde{H}](g) = \frac{1}{n} \text{Tr}[g(\tilde{H})], \quad g \in C(\mathbb{R}).$$

(We will later slightly extend the space of observables.) For example, we can consider the bond energy $\mathcal{D}[\tilde{H}](U_T)$, where $U_T(\epsilon) = \epsilon F_T(\epsilon)$ and $F_T(\epsilon) = (1 + e^{(\epsilon - \mu)/kT})^{-1}$ is the Fermi function. Formally, the value of the observable for the infinite system Ω is the limit of $\mathcal{D}[\tilde{H}](g)$ as $\tilde{\Omega} \uparrow \Omega$.

For future reference we remark that, since H is defined in terms of the lattices R_j and the hopping functions $h_{\alpha\alpha'}$, we will say “ H satisfies Assumptions 2.2 and 2.1” to mean that “ R_1, R_2 satisfy Assumption 2.1 and $h_{\alpha\alpha'}$ satisfy Assumption 2.2”.

2.3. Local Density of States. The next step is to define the local density of states distribution, which will allow us to identify local site contribution to an observable. Consider a finite sub-system $\tilde{\Omega} \subset \Omega$ with associated hamiltonian $\tilde{H} \in M_n(\mathbb{C})$, then the local density of states distribution is defined as

$$\mathcal{D}_k[\tilde{H}](g) = [g(\tilde{H})]_{kk}, \quad k \in \tilde{\Omega}, \quad g \in C(\mathbb{R}).$$

Note that

$$\frac{1}{n} \sum_{k \in \tilde{\Omega}} \mathcal{D}_k[\tilde{H}](g) = \mathcal{D}[\tilde{H}](g).$$

This reformulation puts us very close to the setting of Theorem 2.1. It remains to control the dependence of $\mathcal{D}_k[\tilde{H}](g)$ on $\tilde{\Omega}$, which we will achieve in the next section by fixing k and letting $\tilde{\Omega} \uparrow \Omega$ while controlling the error.

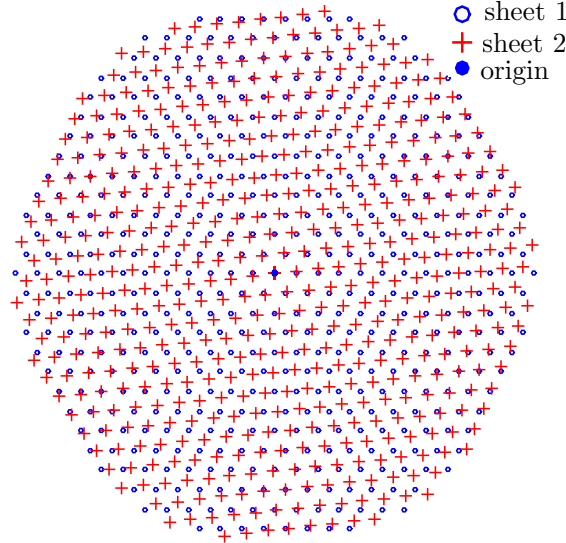


FIGURE 3. All the sites in Ω_r for a hexagonal bravais lattice. The central site for sheet 1 is highlighted.

Towards that end we now specify a sequence of local degree of freedom spaces,

$$\Omega_r = [\mathcal{R}_1 \cap B_r] \times \mathcal{A}_1 \cup [\mathcal{R}_2 \cap B_r] \times \mathcal{A}_2, \quad \text{for } r > 0;$$

see also Figure 3. For $r > 0$ and $b \in \mathbb{R}^2$ we define $H_{r,j}(b) \in M_{|\Omega_r|}(\mathbb{C})$ by

$$[H_{r,j}(b)]_{R\alpha, R'\alpha'} = h_{\alpha\alpha'}(b(\delta_{\alpha \in \mathcal{A}_{P_j}} - \delta_{\alpha' \in \mathcal{A}_{P_j}}) + R - R'),$$

for $R\alpha, R'\alpha' \in \Omega_r$. Physically, $H_{r,j}(b)$ describes a cluster of radius r of the bilayer system in which the sheet P_j is shifted by b . The local configuration is determined

by the relative shift, so b indexes which local configuration we are considering. Then

$$(2.2) \quad \mathcal{D}_\alpha[H_{r,j}(b)] := \mathcal{D}_{0\alpha}[H_{r,j}(b)] \quad \text{for } \alpha \in \mathcal{A}_j$$

is an approximate local density of states distribution of the infinite system at a local configuration indexed by $b \in \Gamma_{P_j}$ at orbital α on sheet j .

2.4. Thermodynamic Limit. We now consider the limit as $r \rightarrow \infty$ of the LDoS, which will allow us to define the DoS for the infinite system. Let

$$E[H] := \sup_{r>0, j \in \{1,2\}} \left[\sup_{b \in \Gamma_j} \|H_{r,j}(b)\|_2 \right] < \infty,$$

where $\|\tilde{H}\|_2 := \sup_{\psi \in \mathbb{C}^n \setminus \{0\}} \|\tilde{H}\psi\|_2 / \|\psi\|_2$, for $\tilde{H} \in M_n(\mathbb{C})$. Then the local density of states distribution will be supported on the interval

$$S[H] = [-E[H], E[H]].$$

We can now generalize observables to be functions $g \in C(S[H])$ and supply this space with the norm

$$\|g\|_\infty := \sup_{x \in S[H]} |g(x)|, \quad \text{for } g \in C(S[H]).$$

For $U \subset \mathbb{C}$, we define the distance

$$d(U, S[H]) = \inf_{z \in U, z' \in S[H]} |z - z'|.$$

This is a bound on the distance between U and the spectrum. To pass to the limit in the LDoS and later in the DoS, we narrow down admissible test functions to

$$\Lambda := \{g \in C(\mathbb{R}) \mid g \text{ is analytic on } S[H]\}.$$

If $g \in \Lambda$, then there exists $\tilde{d} > 0$ such that $g \in \Lambda_{\tilde{d}}$, which is defined as

$$\Lambda_{\tilde{d}} := \{g \in C(\mathbb{R}) \mid g \text{ is analytic at } z \text{ for } d(z, S[H]) \leq \tilde{d}\}.$$

Theorem 2.2. (1) Suppose that H satisfies Assumptions 2.1 and 2.2. Then, for $\alpha \in \mathcal{A}_j$, there exists a function $\mathcal{D}_\alpha[H] : \Gamma_{P_j} \times C(S[H]) \rightarrow \mathbb{C}$ such that, for $g \in \Lambda$,

$$\mathcal{D}_\alpha[H_{r,j}(b)](g) \rightarrow \mathcal{D}_\alpha[H](b, g) \quad \text{as } r \rightarrow \infty.$$

(The distribution $\mathcal{D}_\alpha[H](b, g)$ is the local density of states for the infinite system.)

(2) The map $g \mapsto \mathcal{D}_\alpha[H](b, g)$ is a bounded linear functional, more precisely,

$$|\mathcal{D}_\alpha[H](b, g)| \leq \|g\|_\infty \quad \text{for } g \in C(S[H]).$$

(3) There exist constants $C, \gamma' > 0$ such that, for $\tilde{d} > 0$ and $g \in \Lambda_{\tilde{d}}$,

$$|\mathcal{D}_\alpha[H](b, g) - \mathcal{D}_\alpha[H_{r,j}(b)](g)| \leq C\tilde{d}^{-2} \sup_{d(z, S[H]) < \tilde{d}} |g(z)| e^{-\gamma' \tilde{d} r}.$$

We next analyze the regularity of the map $b \mapsto \mathcal{D}_\alpha[H](b, g)$ for fixed g , which will allow us to integrate with respect to b . Let $n \in \mathbb{Z}$, $m = (m_1, m_2) \in \mathbb{N}^2$ such that $m_1 + m_2 \leq n$. Then, for $f \in C^n(\mathbb{R}^2)$, we employ the usual multi-index notation

$$\partial_m f = \frac{\partial^{m_1+m_2} f}{\partial x_1^{m_1} \partial x_2^{m_2}}.$$

Theorem 2.3. Suppose $h_{\alpha\alpha'} \in C^n(\mathbb{R}^2)$ for $n \in \mathbb{N} \cup \{0, \infty\}$, $\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}$ is uniformly continuous for $m + m' \leq n$ and satisfies

$$|\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}(r)| \leq C e^{-\gamma' r}.$$

Then, for $\alpha \in \mathcal{A}_j$ and $g \in \Lambda$,

$$\mathcal{D}_\alpha[H](\cdot, g) \in C_{\text{per}}^n(\Gamma_{P_j}).$$

Our next objective is to rigorously define the density of states distribution for the infinite incommensurate bilayer system H . Taking a sequence of finite incommensurate clusters surrounded by vacuum that grow towards infinity and combining our results on the equidistribution of local configurations with the convergence of the local density of states we obtain the following representation formula.

Theorem 2.4. Suppose that H satisfies Assumptions 2.1 and 2.2. Then there exists a bounded linear functional $\mathcal{D}[H] : C(S[H]) \rightarrow \mathbb{C}$ such that, for $g \in \Lambda$, we have

$$\mathcal{D}[H_{r,j}(0)](g) \rightarrow \mathcal{D}[H](g) \quad \text{as } r \rightarrow \infty, \quad \text{for } j = 1, 2,$$

and

$$\mathcal{D}[H](g) = \nu \sum_{j=1}^2 \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H](b, g) db,$$

where

$$\nu = \frac{1}{|\mathcal{A}_2| \cdot |\Gamma_1| + |\mathcal{A}_1| \cdot |\Gamma_2|}.$$

If $g \in \Lambda_{\tilde{d}}$, then we have the explicit error bound

$$\left| \mathcal{D}[H](g) - \nu \sum_{j=1}^2 \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H_{r,j}(b)](g) db \right| \leq C \tilde{d}^{-2} \sup_{d(z, S[H]) < \tilde{d}} |g(z)| e^{-\gamma \tilde{d} r},$$

where C, γ are independent of r, \tilde{d} and g .

Remark 2.1. The finite systems employed in the thermodynamic limit are defined by the matrices $H_{r,j}(0)$ for $j = 1, 2$. They represent finite incommensurate clusters surrounded by vacuum. Since the boundary Hamiltonian entries are not chosen by DFT calculations or experimental values they will not be accurate. However, as long as the boundary coefficients satisfy Assumption 2.2, the limit of the density of states $\mathcal{D}[H_{r,j}(0)]$ will be independent of the choice of boundary terms.

Remark 2.2. For the sake of convenience, we have chosen a circular shape for the approximating domains. Weaker requirements can be readily formulated, e.g., domains $\tilde{\Omega}$ should contain balls centered at the origin with radii growing to infinity, while at the same time keeping a suitable bound on the surface area to volume ratio.

Remark 2.3. The Riesz-Markov-Kakutani Representation Theorem states that the dual space of the continuous compact functions are the Radon measures. Since all our density of states and local density of states operators are continuous linear functionals over the space of compact continuous functions, they are all Radon measures.

Remark 2.4. This methodology can easily be extended to three or more incommensurate layers, but at the cost of multiple integrals, since one must integrate over all relative shifts between the layers. The local density of states can be easily analyzed for multiple layers without adding much to the cost.

3. NUMERICAL SIMULATIONS

3.1. Quadrature. To compute the integrals occurring in Theorem 2.4 numerically, we can use the smoothness properties from Theorem 2.3, which can be strengthened further by assuming analyticity on $h_{\alpha\alpha'}$.

Theorem 3.1. *Assume $h_{\alpha\alpha'}$ is analytic and satisfies Assumption 2.2. Let*

$$S_j = \left\{ A_j \left(\frac{i_1/N_{disc}}{i_2/N_{disc}} \right) : 0 \leq i_1, i_2 < N_{disc} \right\}$$

be the uniform discretization sample points. Then we have

$$\begin{aligned} \left| \frac{|\Gamma_{P_j}|}{N_{disc}^2} \sum_{b \in S_{P_j}} \sum_{\alpha \in \mathcal{A}_j} \mathcal{D}_\alpha[H](b, g) - \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H](b, g) db \right| \\ \leq C \tilde{d}^{-1} \sup_{z: d(z, S[H]) < \tilde{d}} |g(z)| e^{-\gamma'' \tilde{d} N_{disc}} \end{aligned}$$

for some $\gamma'' > 0$.

Remark 3.1. *In practice, $h_{\alpha\alpha}$ has a finite cut-off and hence cannot be analytic. However, we can think of it as an approximation to an exact analytic $\bar{h}_{\alpha\alpha}$. Preasymptotically, it is therefore useful to treat $h_{\alpha\alpha'}$ as if it were itself analytic.*

3.2. Kernel Polynomial Method Approximation. A complete eigensolve on $H_{r,j}(b)$ for each quadrature point b is computationally expensive, with scaling $O(r^6)$. Instead we use a Chebyshev Kernel Polynomial Method (KPM) to compute the density of states [19]. This method scales as $O(r^2)$, where the constant depends on the desired accuracy. It yields the density of states operator as a smooth function from which multiple observables can then be computed.

Lemma 3.1. *Assume that H satisfies Assumptions 2.2 and 2.1 and that $f \in C(\mathbb{R} \times \mathbb{R}; \mathbb{C})$ and $g \in \Lambda$; then*

$$\int \mathcal{D}[H](f(\varepsilon, \cdot)) g(\varepsilon) d\varepsilon = \mathcal{D}[H] \left(\int f(\varepsilon, \cdot) g(\varepsilon) d\varepsilon \right).$$

Proof. This result follows immediately from Remark 2.3 and Fubini's Theorem. \square

We note that $|\mathcal{D}[H](g)| \leq \|g\|_\infty$, and hence

$$(3.1) \quad \left| \mathcal{D}[H] \left(\int f(\varepsilon, \cdot) g(\varepsilon) d\varepsilon \right) - \mathcal{D}[H](g) \right| \leq \left\| \int f(\varepsilon, \cdot) g(\varepsilon) d\varepsilon - g \right\|_\infty.$$

Note that this bound trivially extends from Λ to $C(S[H])$. Moreover, if $f(\varepsilon, e) \approx \delta(\varepsilon - e)$, then the smooth function

$$D_f(\varepsilon) := \mathcal{D}[H](f(\varepsilon, \cdot)) \approx \mathcal{D}[H]$$

in the sense of Equation 3.1. We now choose a convenient f .

Recall that the Chebyshev polynomials are a basis defined recursively by

$$(3.2) \quad T_0(e) = 1, \quad T_1(e) = e, \quad \text{and} \quad T_{n+1}(e) = 2eT_n(e) - T_{n-1}(e).$$

The polynomials are orthogonal in the sense that

$$\int_{-1}^1 \frac{1}{\pi \sqrt{1-e^2}} T_n(e) T_m(e) de = \frac{1 + \delta_{0n}}{2} \delta_{nm}.$$

An approximation to the shifted delta function $\delta(e - \varepsilon)$, at $\varepsilon \in (-1, 1)$, is given by

$$\hat{\chi}_p(\varepsilon, e) = \frac{1}{\pi\sqrt{1-\varepsilon^2}} \sum_{m \leq p} g_m^p T_m(\varepsilon) T_m(e), \quad e, \varepsilon \in (-1, 1),$$

where

$$g_m^p = (2 - \delta_{m0}) \frac{(p - m + 1) \cos(\frac{\pi m}{p+1}) + \sin(\frac{\pi m}{p+1}) \arctan(\frac{\pi}{p+1})}{p + 1}$$

are the so-called Jackson coefficients designed to remove the Gibbs phenomenon [19].

To approximate the density of states on the interval $S[H] = [-E[H], E[H]]$, we rescale

$$\chi_p(\varepsilon, e) := \eta \hat{\chi}_p(\eta \varepsilon, \eta e), \quad e, \varepsilon \in (-1/\eta, 1/\eta),$$

where η is a positive constant selected so that $E[H] \leq 1/\eta$.

We approximate $\mathcal{D}[H]$ by

$$D_{\chi_p}(\varepsilon) = \nu \sum_{j=1}^2 \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H](b, \chi_p(\varepsilon, \cdot)) db,$$

and subsequently approximate the integrand $\mathcal{D}_\alpha[H](b, \chi_p(\varepsilon, \cdot))$ by

$$\begin{aligned} \mathcal{D}_\alpha[H_{r,j}(b)](\chi_p(\varepsilon, \cdot)) &= [\chi_p(\varepsilon, H_{r,j}(b))]_{0\alpha, 0\alpha} \\ (3.3) \quad &= \frac{\eta}{\pi\sqrt{1-(\eta\varepsilon)^2}} \sum_{m \leq p} g_m^p T_m(\eta\varepsilon) [\eta T_m(H_{r,j}(b))]_{0\alpha, 0\alpha}. \end{aligned}$$

Note that for all ε , the calculation requires the same $[T_m(\eta H_{r,j}(b))]_{0\alpha, 0\alpha}$ coefficients, which is the core of our **Algorithm A**.

The approximation error for the output $D(\varepsilon)$ of Algorithm A is estimated in the following result.

Theorem 3.2. *Suppose that H satisfies Assumptions 2.1 and 2.2, then for $g \in \Lambda_{\tilde{d}}$,*

$$\begin{aligned} \left| \mathcal{D}[H](g) - \int D(\varepsilon) g(\varepsilon) d\varepsilon \right| &\leq \underbrace{C \tilde{d}^{-2} \sup_{d(z, S[H]) < \tilde{d}} |g(z)| e^{-\gamma \tilde{d} r}}_{\text{Truncation Error}} + \\ &\quad \underbrace{C \tilde{d}^{-1} \sup_{d(z, S[H]) < \tilde{d}} |g(z)| e^{-\gamma' \tilde{d} N_{disc}}}_{\text{Discretization Error}} + \underbrace{C' \left\| g - \int \chi_p(\varepsilon, \cdot) g(\varepsilon) d\varepsilon \right\|_\infty}_{\text{Kernel Polynomial Method Error}}. \end{aligned}$$

Here $\gamma, \gamma' > 0$ are independent of the choice of \tilde{d} .

Proof. The Truncation Error follows from Theorem 2.2, the Discretization Error from Theorem 3.1, and the Kernel Polynomial Error from (3.1). \square

Remark 3.2. *If we do not assume that $h_{\alpha\alpha'}$ is analytic and use $h_{\alpha\alpha'} \in C_0^n(\mathbb{R}^2)$ instead, the Truncation Error above is replaced with the standard periodic discretization error [18, Theorem 1], but the bound does not give the dependence of N_{disc} on \tilde{d} .*

Algorithm A: Approximate DoS

Step 1: Choose quadrature parameter $N_{\text{disc}} \in \mathbb{N}$ and domain truncation radius $r > 0$. For each $j \in \{1, 2\}$ and $b \in S_{P_j}$ construct the matrix $H_{r,j}(b)$.

Step 2: Let $e_i \in \mathbb{R}^{|\Omega_r|}$ such that $[e_i]_j = \delta_{ij}$ is the i^{th} coordinate vector. Using the recursion (3.2) we compute, for $\alpha \in \mathcal{A}_j$,

$$\begin{aligned} v_0 &= e_{0\alpha} \\ v_1 &= \eta H_{r,j}(b) e_{0\alpha} \\ \text{store: } [T_0(\eta H_{r,j}(b))]_{0\alpha,0\alpha} &= e_{0\alpha} \cdot v_0 \text{ and } [T_1(\eta H_{r,j}(b))]_{0\alpha,0\alpha} = e_{0\alpha} \cdot v_1 \\ \text{for loop: } 1 \leq m &\leq p-1 \\ v_{m+1} &= 2\eta H_{r,j}(b) v_m - v_{m-1} \\ \text{store: } [T_{m+1}(\eta H_{r,j}(b))]_{0\alpha,0\alpha} &= e_{0\alpha} \cdot v_{m+1} \end{aligned}$$

This yields the coefficients $[T_m(\eta H_{r,j}(b))]_{0\alpha,0\alpha}$ for (3.3).

Step 3: Compute the expression

$$\mathcal{D}_\alpha[H_{r,j}(b)](\chi_p(\varepsilon, \cdot)) = \frac{\eta}{\pi \sqrt{1 - (\eta\varepsilon)^2}} \sum_{m \leq p} g_m^p T_m(\eta\varepsilon) [T_m(\eta H_{r,j}(b))]_{0\alpha,0\alpha}.$$

This yields a local density of states approximation, which is interesting in its own right.

Step 4: The total density of states approximation is obtained by evaluating

$$D(\varepsilon) := \frac{\nu}{N_{\text{disc}}^2} \sum_{j=1}^2 \sum_{\alpha \in \mathcal{A}_j} \sum_{b \in S_{P_j}} |\Gamma_{P_j}| \cdot \mathcal{D}_\alpha[H_{r,j}(b)](\chi_p(\varepsilon, \cdot))$$

for all desired ε .

3.3. Convergence Rates. We briefly discuss a heuristic to choose the approximation parameters $p, N_{\text{disc}} \in \mathbb{N}$ and $r > 0$. In practice, one is interested in calculating the density of states at a point or in calculating an observable $\mathcal{D}[H](g)$ for $g \in \Lambda_{\tilde{d}}$.

For the first case, we note that χ_p acts similar to an approximation to the identity of width proportional to p^{-1} [19] with well preserved regularity because of the Jackson coefficients. For analytic purposes, we can consider $\chi_p(\varepsilon, e) \sim p^{-1} \phi((\varepsilon - e)/p)$ for some analytic function ϕ , $|\phi(x)| < e^{-c|x|}$ for $x \in S[H]$ and for some $c > 0$. An approximation of the density of states at a given energy point ε is given by $\mathcal{D}[H](\chi_p(\varepsilon, \cdot)) \sim \mathcal{D}[H](p^{-1} \phi((\cdot - \varepsilon)/p))$. To approximate $\mathcal{D}[H](\chi_p(\varepsilon, \cdot))$, we use Theorem 3.2 letting $\tilde{d} \sim p^{-1}$ to see that the errors will be balanced if

$$(3.4) \quad r \sim N_{\text{disc}} \sim p \log(p)$$

Suppose the density of states is a function, i.e.,

$$\mathcal{D}[H](g) = \int \text{DoS}(\epsilon) g(\epsilon) d\epsilon,$$

where DoS has Lipschitz constant M . Then we can estimate

$$|\text{DoS}(\varepsilon) - \mathcal{D}[H](\chi_p(\varepsilon, \cdot))| \leq Mp^{-1}.$$

to obtain

$$|D(\varepsilon) - \text{DoS}(\varepsilon)| \leq C'(pe^{-\gamma' \frac{N_{\text{disc}}}{p}} + p^2 e^{-\gamma \frac{r}{p}} + Mp^{-1}).$$

If the constants in (3.4) are chosen sufficiently small, we have

$$(3.5) \quad |D(\varepsilon) - \text{DoS}(\varepsilon)| \leq (M + C)p^{-1},$$

where $C > 0$ is independent of smoothness properties of DoS.

If the DoS is C^2 at a point ε of interest, then we may even expect

$$(3.6) \quad |D(\varepsilon) - \text{DoS}(\varepsilon)| \leq Cp^{-2},$$

due to the fact that $\int xe^{-ax^2} dx = 0$ for any $a > 0$.

For the second case, when the observable $g \in \Lambda$ is fixed (no polynomial degree approximation parameter p), we have *in principle* exponential decay of the error in r and N_{disc} . This seems to imply that it would be optimal to calculate the observable directly using an eigensolve, thus avoiding the slower decay in p . However the decay rate in r is strongly coupled to the value of \tilde{d} from Theorem 2.2, which is fairly small for interesting observables. Therefore, the involved matrices are typically quite large, rendering direct eigensolves impractical.

3.4. Numerical Results. We test our approximation scheme using a tight-binding model for twisted bilayer graphene [5] with a relative twist angle of 6° . We fix an $\alpha \in \mathcal{A}_1$ and then verify numerically the following two results:

- (1) As predicted in Theorem 2.2, $\mathcal{D}_\alpha[H_{r,1}(b)](\chi_p(\varepsilon, \cdot)) \rightarrow \mathcal{D}_\alpha[H](b, \chi_p(\varepsilon, \cdot))$ as $r \rightarrow \infty$ with exponential rate: see Figure 4.
- (2) As predicted by Theorem 3.2 and (3.6), $D \rightarrow \text{DoS}$ pointwise as $p, r, N_{\text{disc}} \rightarrow \infty$, with quadratic rate: see Figure 5.

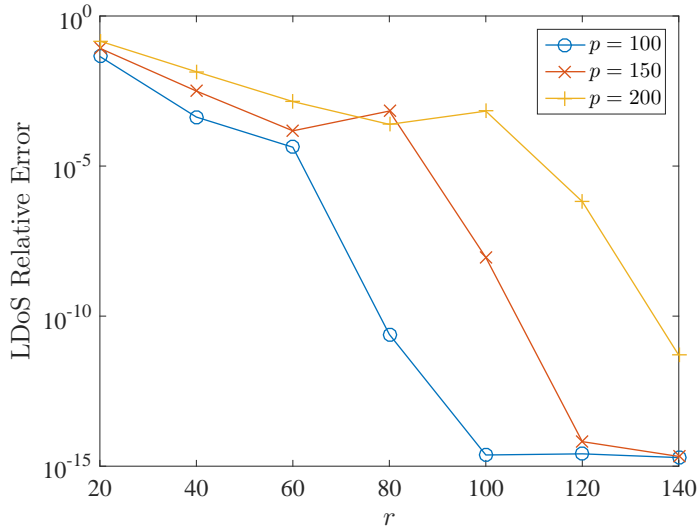


FIGURE 4. Relative error of $\mathcal{D}_\alpha[H_{r,1}(0)](\chi_p(0, \cdot))$ converging to $\mathcal{D}_\alpha[H](\chi_p(0, \cdot))$, for increasing values of p .

Furthermore, we demonstrate the practicality of Algorithm A by reproducing twisted bilayer effects in the density of states of two stacked graphene sheets with

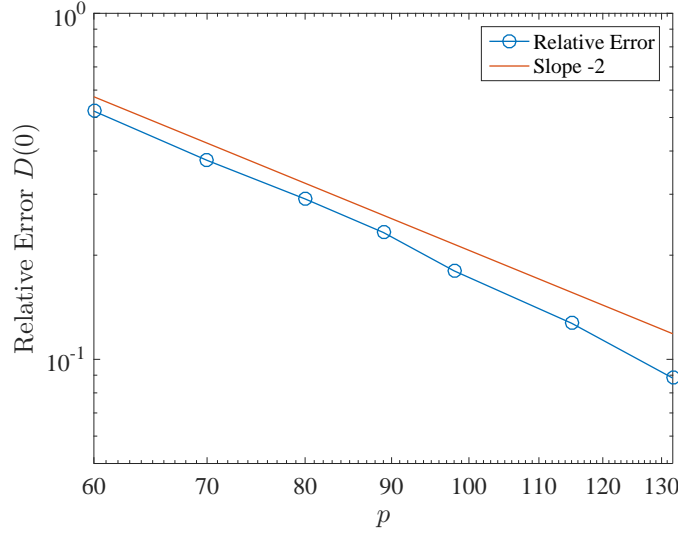


FIGURE 5. Relative error of $D(0) \rightarrow \text{DoS}(0)$ pointwise, where r and N_{disc} scale as in (3.4). The slope is $-1.98 \approx -2$, as predicted in (3.6).

a relative twist of 6° as predicted in [5] (See Figure 6). We included the DoS for monolayer graphene for comparison. The conical region near the -0.6 energy region is called the Dirac cone. When the two layers interact, the curve splits near the cone tip (the Dirac point) forming two Van Hove Singularities on either side of the tip. In practice the VHS needs higher resolution. We will explore how to achieve high resolutions in a future work.

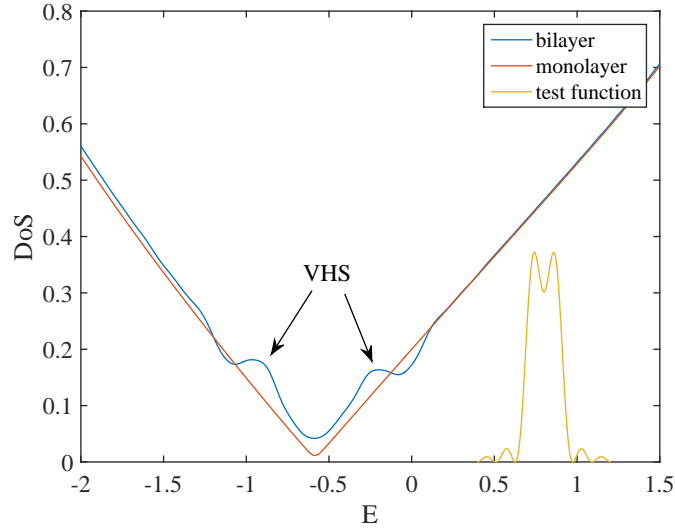


FIGURE 6. Approximation of the DoS with $r = 180$, $p = 700$, and $N_{\text{disc}} = 4$. We can see Van Hove Singularities (VHS) forming near the Dirac Point, agreeing with theoretical results [5]. We include the test function, which is to scale in the E-axis, but not in the DoS-axis.

4. PROOFS

To attain bounds on the density of states objects, we will use resolvent bounds as introduced in [3]. We denote \mathcal{C} a contour around $S[H]$, which contains the spectrum. We can write for $\tilde{\Omega} \subset \Omega$ finite, $\tilde{H} \in M_{|\tilde{\Omega}|}(\mathbb{C})$, $k \in \Omega$, and g analytic

$$[g(H_{\tilde{\Omega}})]_{kk} = \frac{1}{2\pi i} \oint_{\mathcal{C}} g(z) [(z - \tilde{H})^{-1}]_{kk} dz.$$

We will then rely on decay estimates for $[(z - \tilde{H})^{-1}]_{kk}$ as $\tilde{\Omega} \uparrow \Omega$. We will vary our choice of \mathcal{C} to tune the error bounds.

4.1. Proof of Theorem 2.1. Although this result is conceptually close to the equidistribution theorem [20], our specific statement of the result seems to be unavailable. Hence we prefer to give a complete proof. Without loss of generality, we let $j = 1$ and hence $P_j = 2$. Then we wish to show for $g \in C_{\text{per}}(\Gamma_2)$, we have

$$\frac{1}{\#\mathcal{R}_1 \cap B_r} \sum_{\ell \in \mathcal{R}_1 \cap B_r} g(\ell) \rightarrow \frac{1}{|\Gamma_2|} \int_{\Gamma_2} g(b) db.$$

Upon transforming coordinates we may assume without loss of generality that $A_1 = \text{Id}$. Hence for some matrix A dependent on the original coordinates and $V_r = |AB_r|$ we get

$$(4.1) \quad \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} g(n) \rightarrow \frac{1}{|\Gamma_2|} \int_{\Gamma_2} g(x) dx.$$

Since $C_{\text{per}}^\infty(\Gamma_2)$ is dense in $C_{\text{per}}(\Gamma_2)$, we assume $g \in C_{\text{per}}^\infty(\Gamma_2)$. On expanding g into Fourier modes, it suffices to show (4.1) for an arbitrary fourier mode $g(x) = e^{2\pi i m \cdot A_2^{-1} x}$ where $m \in \mathbb{Z}^2$.

If $m = (0, 0)$, then the left-hand side of (4.1) converges to 1, which is the value of the right-hand side.

For $m \neq (0, 0)$, the left-hand side of (4.1) vanishes, so we need to prove that $\frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} f(n) \rightarrow 0$ as $r \rightarrow \infty$. We first rewrite

$$\frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} f(n) = \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} e^{2\pi i m^t A_2^{-1} n} = \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} e^{2\pi i a \cdot n},$$

where $(a_1, a_2) = m^t A_2^{-1}$. If both a_1 and a_2 were rational, then this would contradict Assumption 2.1. Hence we assume, without loss of generality, that $a_2 \notin \mathbb{Q}$.

Let $c > 0$ such that

$$n \in \mathbb{Z}^2 \cap AB_r \Rightarrow n_1 \in [-cr, cr].$$

Moreover, for $n_1 \in [-cr, cr] \cap \mathbb{Z}$ let $f_1(n_1), f_2(n_2) \in \mathbb{Z}$ such that $(n_1, n_2) \in \mathbb{Z}^2 \cap AB_r$ if and only if $f_1(n_1) \leq n_2 \leq f_2(n_2)$.

We can now compute

$$\begin{aligned} \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} e^{2\pi i a \cdot n} &= \frac{1}{V_r} \sum_{n_1 \in [-cr, cr] \cap \mathbb{Z}} e^{2\pi i a_1 n_1} \sum_{n_2 = f_1(n_1)}^{f_2(n_2)} e^{2\pi i a_2 n_2} \\ &= \frac{1}{V_r} \sum_{n_1 \in [-cr, cr] \cap \mathbb{Z}} e^{2\pi i a_1 n_1} \frac{e^{2\pi i a_2 (f_1(n_1)+1)} - e^{2\pi i a_2 (f_2(n_1)+1)}}{1 - e^{2\pi i a_2}}. \end{aligned}$$

Since a_2 is irrational, $1 - e^{2\pi i a_2} \neq 0$, hence we can estimate

$$\left| \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \cap AB_r} e^{2\pi i a \cdot n} \right| \leq \frac{4cr}{|1 - e^{2\pi i a_2}| V_r} \leq Cr^{-1},$$

which vanished in the limit $r \rightarrow \infty$, as required. This completes the proof of Theorem 2.1.

4.2. Proof of Theorem 2.2. Recall that

$$\Lambda := \{g \in C(\mathbb{R}) \mid g \text{ is analytic on } S[H]\}.$$

In particular, note that Λ is dense in $C(S[H])$, in the sense that for any $f \in C(S[H])$ and $\epsilon > 0$, there exists $g \in \Lambda$ such that

$$\|g|_{S[H]} - f\|_\infty < \epsilon.$$

This will be useful for extending the density of states operators from Λ to $C(S[H])$.

Lemma 4.1. *Suppose $\tilde{H} \in M_n(\mathbb{C})$, and $y : \{1, 2, \dots, n\} \rightarrow \mathbb{R}^2$ such that*

$$|\tilde{H}_{k\ell}| \leq C e^{-\tilde{\gamma}|y(k) - y(\ell)|}$$

for some $\tilde{\gamma} > 0$. Let $N \in \mathbb{N}$, $r' > 0$ and suppose that for all $x \in \mathbb{R}^2$ $|\#\{y(j) : y(j) \in B_{r'}(x)\}| < N$. Then there exists $\gamma > 0$ such that, for all $z \in \mathbb{C}$, $\text{dist}(z, S[H]) \geq \tilde{d}$,

$$\left| [(z - \tilde{H})^{-1}]_{k\ell} \right| \leq C' \tilde{d}^{-1} e^{-\gamma \tilde{d}|y(k) - y(\ell)|}$$

Here C' and γ are dependent on $\tilde{\gamma}, N, r'$, and C .

Proof. This is a version of Lemma 2.2 from [3]. □

In particular, the previous lemma applies to the matrices $H_{r,j}(b)$. To apply it we will set $y = \mathfrak{R}$ where in the following we define

$$\mathfrak{R} : \Omega \rightarrow \mathbb{R}^2, \quad \mathfrak{R}(R\alpha) = R.$$

For the next lemma, recall the definition of $H_{r',j}(b)$ from (2.2).

Lemma 4.2. *Suppose that H satisfies Assumptions 2.2 and 2.1. Let $\tilde{\Omega} \subset \Omega$ be a set of indices and $\tilde{H}_j(b)$ be the matrix defined over $\tilde{\Omega}$ with shift b relative to sheet j , that is,*

$$[\tilde{H}_j(b)]_{R\alpha, R'\alpha'} = h_{\alpha\alpha'}(b(\delta_{\alpha \in \mathcal{A}_{P_j}} - \delta_{\alpha' \in \mathcal{A}_{P_j}}) + R - R').$$

Suppose that $r' > 0$ such that $\Omega_{r'} \subset \tilde{\Omega}$ and $\tilde{d} > 0$ such that $d(z, S[H]) > \tilde{d}$, then

$$\begin{aligned} & \left| [(z - \tilde{H}_j(b))^{-1}]_{k\ell} - [(z - H_{r',j}(b))^{-1}]_{k\ell} \right| \\ & \leq C \tilde{d}^{-2} \min \left\{ e^{-\gamma \tilde{d}|\mathfrak{R}(k) - \mathfrak{R}(\ell)|}, r' e^{-\gamma \tilde{d} \min\{r' - |\mathfrak{R}(k)|, r' - |\mathfrak{R}(\ell)|\}} \right\}, \end{aligned}$$

where C and γ are independent of $\tilde{\Omega}$ and r' (See Figure 7).

Proof. We define the matrix $\tilde{H}_j^{r'}(b) \in M_{|\tilde{\Omega}|}(\mathbb{C})$ such that

$$[\tilde{H}_j^{r'}(b)]_{k\ell} = \begin{cases} H_{r',j}(b) & \text{if } k, \ell \in \Omega_{r'} \\ 0 & \text{otherwise} \end{cases}.$$

We write $\tilde{H}_j(b) = \tilde{H}_j^{r'}(b) + (\tilde{H}_j(b) - \tilde{H}_j^{r'}(b))$, and

$$[(z - \tilde{H}_j(b))^{-1}]_{k\ell} = [(z - \tilde{H}_j^{r'}(b) - (\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)))^{-1}]_{k\ell}.$$

Thus, after defining

$$B(\lambda) = z - \tilde{H}_j^{r'}(b) - \tilde{\lambda}(\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)),$$

and

$$f(\lambda) = [B(\lambda)^{-1}]_{k\ell}$$

we need to estimate $f(1) - f(0)$. Differentiating with respect to λ yields

$$\begin{aligned} f'(\lambda) &= [B(\lambda)^{-1}(\tilde{H}_j(b) - \tilde{H}_j^{r'}(b))B(\lambda)^{-1}]_{k\ell} \\ &= \sum_{t,s \in \tilde{\Omega}} [B(\lambda)^{-1}]_{kt} [\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)]_{ts} [B(\lambda)^{-1}]_{s\ell}. \end{aligned}$$

Now $[\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)]_{ts}$ is only nonzero if t or $s \notin \Omega_{r'}$. We use the definition

$$\tilde{\Omega} \setminus \Omega_{r'} := \{x : x \in \tilde{\Omega}, x \notin \Omega_{r'}\}.$$

From Lemma 4.1, we have

$$|\tilde{H}(\lambda)^{-1}|_{st} \leq C\tilde{d}^{-1}e^{-\gamma\tilde{d}|\Re(s)-\Re(t)|}.$$

Therefore, we obtain the bound

$$\begin{aligned} |f'(\lambda)| &\leq \sum_{t \in \tilde{\Omega}} \sum_{s \in \tilde{\Omega} \setminus \Omega_{r'}} |[B(\lambda)^{-1}]_{kt} [\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)]_{ts} [B(\lambda)^{-1}]_{s\ell}| \\ &\quad + \sum_{s \in \tilde{\Omega}} \sum_{t \in \tilde{\Omega} \setminus \Omega_{r'}} |[B(\lambda)^{-1}]_{kt} [\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)]_{ts} [B(\lambda)^{-1}]_{s\ell}| \\ &\leq C\tilde{d}^{-2} \sum_{t \in \tilde{\Omega}} \sum_{s \in \tilde{\Omega} \setminus \Omega_{r'}} e^{-\gamma\tilde{d}(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)} \\ &\quad + C\tilde{d}^{-2} \sum_{s \in \tilde{\Omega}} \sum_{t \in \tilde{\Omega} \setminus \Omega_{r'}} e^{-\gamma\tilde{d}(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)} \\ &\leq C'\tilde{d}^{-2} \min\{e^{-\gamma\tilde{d}|\Re(k)-\Re(\ell)|}, r'e^{-\gamma\tilde{d}\min\{r'-|\Re(k)|, r'-|\Re(\ell)|\}}\}. \end{aligned}$$

Hence, we conclude that

$$\begin{aligned} |[(z - \tilde{H}_j(b))^{-1}]_{k\ell} - [(z - H_{r',j}(b))^{-1}]_{k\ell}| &\leq |f(1) - f(0)| \leq \int_0^1 |f'(\lambda)| d\lambda \\ &\leq C'\tilde{d}^{-2} \min\{e^{-\gamma\tilde{d}|\Re(k)-\Re(\ell)|}, r'e^{-\gamma\tilde{d}\min\{r'-|\Re(k)|, r'-|\Re(\ell)|\}}\}. \quad \square \end{aligned}$$

Lemma 4.2 shows that the resolvent difference is bounded by the site distances from the edge of the first cut-off region (the circle with radius r') and the distance between the two sites. This is consistent with Lemma 4.1.

Let \mathcal{C} be a contour around $S[H]$ such that $\tilde{d}/2 < d(\mathcal{C}, S[H]) < \tilde{d}$. By Lemma 4.2, we have for $g \in \Lambda_{\tilde{d}}$ that

$$\begin{aligned} &|\mathcal{D}_\alpha[H_{r,j}(b)](g) - \mathcal{D}_\alpha[H_{r',j}(b)](g)| \\ &= \left| \frac{1}{2\pi i} \oint_{\mathcal{C}} g(z) \left([(z - H_{r,j}(b))^{-1}]_{0\alpha,0\alpha} - [(z - H_{r',j}(b))^{-1}]_{0\alpha,0\alpha} \right) \right| \\ &\leq C'\tilde{d}^{-2}r' \sup_{z \in \mathcal{C}} |g(z)| e^{-\gamma\tilde{d}r'}. \end{aligned}$$

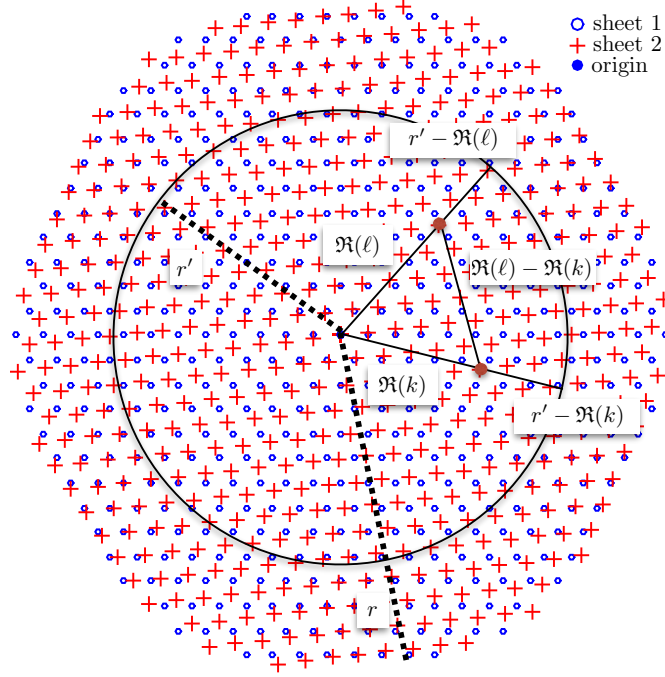


FIGURE 7. For given sites ℓ and k , we plot the relevant distances in solid lines and system radii in dotted lines for considering resolvent error in Lemma 4.2.

Hence $\{\mathcal{D}_\alpha[H_{r_n,j}(b)]\}_n$ is a Cauchy sequence for $r_n \rightarrow \infty$, which therefore has some limit $\mathcal{D}_\alpha[H](b, g)$. $\mathcal{D}_\alpha[H]$ is linear in g , since each element of the Cauchy sequence is linear. Further, we have the error bound

$$|\mathcal{D}_\alpha[H](b, g) - \mathcal{D}_\alpha[H_{r,j}(b)](g)| \leq C' \tilde{d}^{-2} r \sup_{z \in \mathcal{C}} |g(z)| e^{-\gamma \tilde{d} r}.$$

Since the linear functional $\mathcal{D}_\alpha[H_{r,j}(b)]$ is bounded by $\|\mathcal{D}_\alpha[H_{r,j}(b)]\| \leq 1$ we also obtain that $\mathcal{D}_\alpha[H](b, \cdot)$ is a bounded linear functional, and so has a unique extension to a bounded linear functional on the space $C(S[H])$.

This completes the proof of Theorem 2.2.

4.3. Proof of Theorem 2.3.

Lemma 4.3. *Suppose $h_{\alpha\alpha'} \in C^n(\mathbb{R}^2)$ for $n \in \mathbb{N} \cup \{\infty\}$ and $\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}$ is uniformly continuous for $m + m' \leq n$. We further assume the decay estimate*

$$(4.2) \quad |\partial_{b_m} \partial_{b_{m'}} h_{\alpha\alpha'}(r)| \leq C e^{-\gamma' r}.$$

Then for $k = 0\alpha$, we have $b \mapsto [(z - H_{r,j}(b))^{-1}]_{kk} \in C_{\text{per}}^n(\Gamma_{P_j})$, and we have the limit

$$b \mapsto \lim_{r \rightarrow \infty} [(z - H_{r,j}(b))^{-1}]_{kk} \in C_{\text{per}}^n(\Gamma_{P_j})$$

for $d(\{z\}, S[H]) > 0$. Furthermore, for all $b \in \mathbb{R}^2$, $z \mapsto [(z - H_{r,j}(b))^{-1}]_{kk}$ is analytic in $\mathbb{C} \setminus S[H]$.

Proof. We will only consider the derivative ∂_{b_1} ; the treatment of higher (and lower) order derivatives follow the same line of argument, but are more cumbersome. Let

$k = 0\alpha$ for some $\alpha \in \mathcal{A}_j$, then

$$\begin{aligned}\partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk} &= \partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk} \\ &= \sum_{s, \ell \in \Omega_r} [(z - H_{r,j}(b))^{-1}]_{ks} [\partial_{b_1} H_{r,j}(b)]_{s\ell} [(z - H_{r,j}(b))^{-1}]_{\ell k}.\end{aligned}$$

Lemma 4.2 implies that, for $r > r' > 0$,

$$\begin{aligned}R(r, r', k, s) &:= \left| [(z - H_{r,j}(b))^{-1}]_{ks} - [(z - H_{r',j}(b))^{-1}]_{ks} \right| \\ &\leq C \min\{e^{-\gamma|\Re(k) - \Re(s)|}, r' e^{-\gamma \min\{r' - |\Re(k)|, r' - |\Re(s)|\}}\},\end{aligned}$$

where C and γ are independent of r . Note also that, for $s, \ell \in \Omega_{r'}$, we have

$$\partial_{b_1}[H_{r,j}(b)]_{s\ell} = \partial_{b_1}[H_{r',j}(b)]_{s\ell}.$$

Recalling that $\Re(k) = \Re(0\alpha) = 0$, and employing (4.2), we estimate

$$\begin{aligned}& \left| \partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk} - \partial_{b_1}[(z - H_{r',j}(b))^{-1}]_{kk} \right| \\ & \leq C \left(\sum_{s, \ell \in \Omega_{r'}} (R(r, r', k, s) e^{-\gamma|\Re(\ell) - \Re(k)|} + R(r, r', \ell, k) e^{-\gamma|\Re(s) - \Re(k)|}) |\partial_{b_1}[H_{r,j}(b)]_{s\ell}| \right. \\ & \quad \left. + \sum_{s \in \Omega_r, \ell \in \Omega_r \setminus \Omega_{r'}} \left| [(z - H_{r,j}(b))^{-1}]_{ks} [\partial_{b_1} H_{r,j}(b)]_{s\ell} [(z - H_{r,j}(b))^{-1}]_{\ell k} \right| \right) \\ & \leq C' \left(\sum_{s, \ell \in \Omega_{r'}} (R(r, r', k, s) e^{-\gamma|\Re(\ell)|} + R(r, r', \ell, k) e^{-\gamma|\Re(s)|}) e^{-\gamma'|\Re(s) - \Re(\ell)|} + r' e^{-\gamma r'} \right) \\ & \leq C'' r' \sum_{s, \ell \in \Omega_{r'}} e^{-\gamma(r' - |\Re(s)|) - \gamma|\Re(\ell)| - \gamma'|\Re(s) - \Re(\ell)|} + C' r' e^{-\gamma r'} \\ & \leq C''' r'^2 \sum_{s \in \Omega_{r'}} e^{-\gamma(r' - |\Re(s)|) - \min\{\gamma, \gamma'\}|\Re(s)|} + C' r' e^{-\gamma r'} \\ & \leq \tilde{C} e^{-\gamma'' r'},\end{aligned}$$

for any choice of $\gamma'' < \min\{\gamma, \gamma'\}$, where \tilde{C} depends on the choice of γ'' .

Therefore, as $r_n \rightarrow \infty$, $[(z - H_{r_n,j}(b))^{-1}]_{kk}$ forms a Cauchy sequence, and in particular has a limit

$$L_1(b) := \lim_{r \uparrow \infty} \partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk}.$$

Next, we define

$$L(b) := \lim_{r \uparrow \infty} [(z - H_{r,j}(b))^{-1}]_{kk}.$$

We need to show that $\partial_{b_1} L$ exists and satisfies

$$\partial_{b_1} L = L_1.$$

We denote

$$\text{Res}(b) = [(z - H_{r,j}(b))^{-1}]_{kk}.$$

Since $\partial_{b_1} h_{\alpha\alpha'}$ is uniformly continuous there exists a modulus of continuity ω such that $|\partial_{b_1} h(b) - \partial_{b_1} h(b')| \leq \omega(|b - b'|)$. We then observe that, for $\epsilon > 0$ and $e_1 = (1, 0)$,

$$\begin{aligned}\frac{1}{\epsilon} \left(\text{Res}(b + \epsilon e_1) - \text{Res}(b) \right) &= [(z - H_{r,j}(b))^{-1} (\partial_{b_1} H_{r,j}(b) + O(\omega(\epsilon))) (z - H_{r,j}(b))^{-1}]_{kk} \\ &= \partial_{b_1} \text{Res}(b) + O(\omega(\epsilon)).\end{aligned}$$

Here $O(\omega(\epsilon))$ is independent of r . Letting $r \rightarrow \infty$, we have

$$\frac{1}{\epsilon} \left([L(b + \epsilon e_1) - L(b)] \right) = L_1(b) + O(\omega(\epsilon)).$$

Letting $\epsilon \rightarrow 0$ shows that $L \in C_{\text{per}}^{(1,0)}(\Gamma_j)$ and $\partial_{b_1} L = L_1$, which is the desired result.

Continuity with respect to b follows the same argument. Analyticity with respect to z follows from Section 5.2 of [7]. \square

Theorem 2.3 follows immediately from Lemma 4.3.

4.4. Proof of Theorem 2.4. Without loss of generality, let $j = 1$. Fix $g \in \Lambda$, $r > 0$ and $\eta < 1$. Then we have

$$\begin{aligned} \mathcal{D}[H_{r,1}(0)](g) &= \frac{1}{|\Omega_r|} \sum_{k \in \Omega_r} \mathcal{D}_k[H_{r,1}(0)](g) \\ &= \frac{1}{|\Omega_r|} \left(\sum_{k \in \Omega_r \setminus \Omega_{\eta r}} \mathcal{D}_k[H_{r,1}(0)](g) + \sum_{k \in \Omega_{\eta r}} \mathcal{D}_k[H_{r,1}(0)](g) \right). \end{aligned}$$

We define $\mathfrak{A} : \Omega \rightarrow \mathcal{A}_1 \cup \mathcal{A}_2$ such that $\mathfrak{A}(R\alpha) = \alpha$. By Lemma 4.2, we have for $k = R\alpha \in \Omega_{\eta r}$ and $\alpha \in \mathcal{A}_j$ that

$$|\mathcal{D}_k[H_{r,1}(0)](g) - \mathcal{D}_\alpha[H](\text{mod}_{P_j} \circ \mathfrak{R}(k), g)| \leq C \sup_{z \in \mathcal{C}} |g(z)| e^{-\gamma r(1-\eta)}.$$

The site k is at least a distance $r(1 - \eta)$ from the boundary of Ω_r .

Consider the distribution

$$\mathcal{D}[H](g) = \nu \sum_{j=1}^2 \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} D_\alpha[H](b, g) db.$$

Since the integrand is continuous with respect to b (see Theorem 2.3) the integration is well-defined. We now estimate

$$\begin{aligned} |\mathcal{D}[H](g) - \mathcal{D}[H_{r,1}(0)](g)| &\leq \left| \frac{1}{|\Omega_r|} \sum_{k \in \Omega_r \setminus \Omega_{\eta r}} \mathcal{D}_k[H_{r,1}(0)](g) \right| \\ &\quad + \left| \mathcal{D}[H](g) - \frac{1}{|\Omega_{\eta r}|} \sum_{j=1}^2 \sum_{R\alpha \in \Omega_{\eta r} : \alpha \in \mathcal{A}_j} \mathcal{D}_\alpha[H](\text{mod}_{P_j}(R), g) \right| \\ &\quad + \left| \frac{1}{|\Omega_{\eta r}|} \sum_{j=1}^2 \sum_{R\alpha \in \Omega_{\eta r} : \alpha \in \mathcal{A}_j} \mathcal{D}_\alpha[H](\text{mod}_{P_j}(R), g) \right. \\ &\quad \quad \left. - \frac{1}{|\Omega_{\eta r}|} \sum_{k \in \Omega_{\eta r}} \mathcal{D}_k[H_{r,1}(0)](g) \right| \\ &\quad + \left(1 - \frac{|\Omega_{\eta r}|}{|\Omega_r|} \right) \frac{1}{|\Omega_{\eta r}|} \left| \sum_{k \in \Omega_{\eta r}} \mathcal{D}_k[H_{r,1}(0)](g) \right|. \end{aligned}$$

The first and fourth terms are easily seen to be bounded by $O(1 - \eta^2)$. By Theorem 2.1, the second term converges to 0 as $r \rightarrow \infty$. Finally, the third term can be

estimated by

$$\left| \frac{1}{|\Omega_{\eta r}|} \sum_{j=1}^2 \sum_{R\alpha \in \Omega_{\eta r}: \alpha \in \mathcal{A}_j} \mathcal{D}_\alpha[H](\text{mod}_{P_j}(R), g) - \frac{1}{|\Omega_{\eta r}|} \sum_{R\alpha \in \Omega_{\eta r}} \mathcal{D}_{R\alpha}[H_{r,1}(0)](g) \right| \leq C \sup_{z \in \mathcal{C}} |g(z)| e^{-\gamma r(1-\eta)}.$$

Therefore if we choose a pair of sequences $(\eta_j), (r_j)$ such that $\eta_j \uparrow 1$, $r_j \uparrow \infty$, and $r_j(1 - \eta_j) \rightarrow \infty$, we conclude that

$$\mathcal{D}[H_{r,1}(0)](g) \rightarrow \mathcal{D}[H](g).$$

Since $\mathcal{D}[H]$ is a bounded linear functional, it can be extended as before to be a bounded linear functional over $C(S[H])$.

4.5. Proof of Theorem 3.1. We denote $\tilde{z} = (\tilde{z}_1, \tilde{z}_2) \in \mathbb{C}^2$. Let $z \in \mathbb{C}$. Then if $c > 0$ is sufficiently small and $\text{Im}(\tilde{z}_1), \text{Im}(\tilde{z}_2) \in (-c, c)$, we have

$$\|z - H_{r,j}(\tilde{z})\|_2 > 0,$$

and hence $\oint_{\mathcal{C}} g(z) [(z - H_{r,j}(\tilde{z}))^{-1}]_{0\alpha, 0\alpha}$ is analytic at \tilde{z} satisfying $\text{Im}(\tilde{z}_1), \text{Im}(\tilde{z}_2) \in (-c, c)$. We pick a contour \mathcal{C} enclosing $S[H]$ such that $\tilde{d}/2 < d(\mathcal{C}, S[H]) < \tilde{d}$ and then chose $c > 0$ small enough, but keeping $c \sim \tilde{d}$. Since $\oint_{\mathcal{C}} g(z) [(z - H_{r,j}(\tilde{z}))^{-1}]_{0\alpha, 0\alpha} dz$ is analytic with respect to \tilde{z} , we can apply Theorem 2 of [18] to deduce

$$\left| \oint_{\mathcal{C}} g(z) [(z - H_{r,j}(\tilde{z}))^{-1}]_{0\alpha, 0\alpha} \right| < C \tilde{d}^{-1} \sup_{z: d(z, S[H]) < \tilde{d}} |g(z)| e^{-\gamma'' \tilde{d} N_{\text{disc}}}$$

for some $C > 0$ independent of r . The result follows.

5. CONCLUSION

The main result of this work, Theorem 2.4, is a representation formula for the thermodynamic limit of the electronic structure of incommensurate layered heterostructures. The result is reminiscent of Bellissard's noncommutative Brillouin Zone for aperiodic solids [1], replacing on-site randomness with a number-theoretic equidistribution theorem.

Crucially, our representation formula lends itself to numerical approximation. In § 3 we formulate, and analyze at a heuristic level, an efficient kernel polynomial method to approximately compute the density of states in twisted bilayer graphene. This preliminary exploration provides not only quantitative confirmation of our analytical results, but also demonstrates the utility of our approach for applications to real material models.

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